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PHYSICAL AND CHEMICAL PROCESS IN FLAMES

(AFOSR Grant No. FA9550-04-1-0003)

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SUMMARY/OVERVIEW

The objectives of the present program are to develop detailed and simplified chemical kinetics models for hydrocarbon combustion, and to understand and quantify the dynamics of flames. During the reporting period progress were made in the following projects: (1) Skeletal reduction using sensitivity analysis aided by directed relation graph; (2) derivation of analytical solution of quasi-steady state species using directed graph; (3) derivation of a reduced mechanism for lean premixed methane/air flames; (4) extinction and stability analyses of diffusion flames with radiation heat loss; and (5) study of the response of premixed flames to velocity fluctuations with flame stretch effects.

TECHNICAL DISCUSSION

1. Skeletal Reduction with Directed Relation Graph Aided Sensitivity Analysis (DRGASA)

The method of DRG (directed relation graph) was developed in our previous work to reduce large detailed mechanisms to smaller skeletal mechanisms with linear-time algorithm and user-specified accuracy. In this year, the DRG was further extended. First, the conditions for DRG to be applicable was formally studied and it was found that the method is valid for detailed mechanisms for which detailed balancing is honored, which is true for most practical mechanisms of hydrogen and hydrocarbon fuels. Second, DRG was combined with sensitivity analysis to obtain skeletal mechanisms of minimum size for given accuracy requirement.

Specifically, we note that while in many situations DRG reduction can already provide sufficiently compact skeletal mechanisms that can be either directly applied in simulations, the skeletal mechanisms from DRG are frequently not minimal. This is because DRG retains a species if it has a non-negligible effect on any species in the skeletal mechanism, and that not every species in the skeletal mechanism is of equal importance to the major species and global parameters. Therefore it is possible to further eliminate the species which have only minor effects on the major species or global parameters, even though it might have more significant effect on other species which are not of major interests. The identification of the entire set of such eliminable species is nevertheless not straightforward, considering that the induced error by elimination is not small and the coupling of the species is highly nonlinear. A simple and reliable method to eliminate such species has thus been developed by computing the worst-case induced error by the elimination of each species, one at a time. This approach is similar to sensitivity analysis, except it directly evaluates the effects of species elimination instead of using perturbations of the species concentrations. This reduction process is more CPU-time demanding than DRG due to the evaluation of global sensitivities. However, the procedure appears to be

straightforward, reliable, and executable for mechanisms of moderate sizes obtained by DRG reduction.
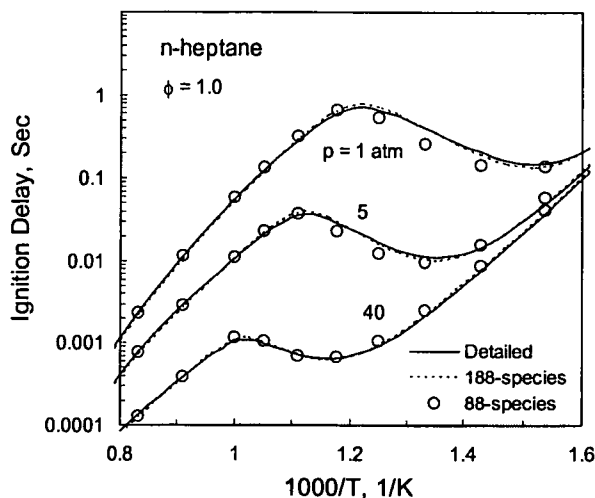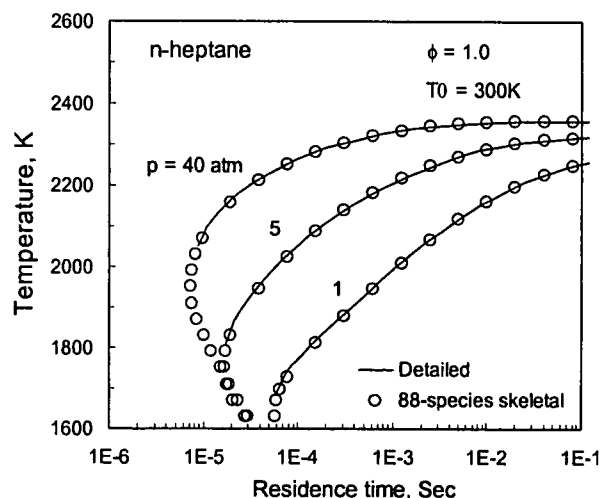

Figure 1

Figure 2

The method of DRGASA developed employs an iterative procedure. In each loop of the iteration, the information obtained from a DRG analysis is exploited to facilitate the subsequent process of sensitivity analysis. Species found to be unimportant by DRG can be directly eliminated, and species found to be highly important by DRG are directly retained. Only the species which are uncertain for DRG need to be tested by sensitivity analysis. As such, the number of species for sensitivity test is significantly reduced. This procedure can be repeated until there is no species further eliminable, and the resulting skeletal mechanism is therefore the minimal.

The method of DRGASA was applied to further reduce the skeletal mechanism of heptane with 188 species and 939 reactions. An 88-species mechanism with 384 reactions was obtained. The skeletal mechanism was validated against the 188-species and detailed mechanism for both ignition processes in auto-ignition and extinction in perfectly stirred reactor (PSR). Figures 1 and 2 show that, while there is significant reduction in mechanism size, only slightly increased error is observed in ignition delays near NTC regime.

2. Analytical Solution of Quasi-Steady State equations with Directed Graph (QSSDG)

A systematic approach was developed to obtain analytical solutions for reduced mechanisms based on QSS assumptions. The method approximates the set of nonlinear algebraic equations for QSS species concentrations with a set of linear equations with good accuracy. This is based on the observation that QSS species are typically low in concentrations, and therefore the reaction rates involving two trace species are likely to be small compared with those of the rate-controlling reactions. The linear
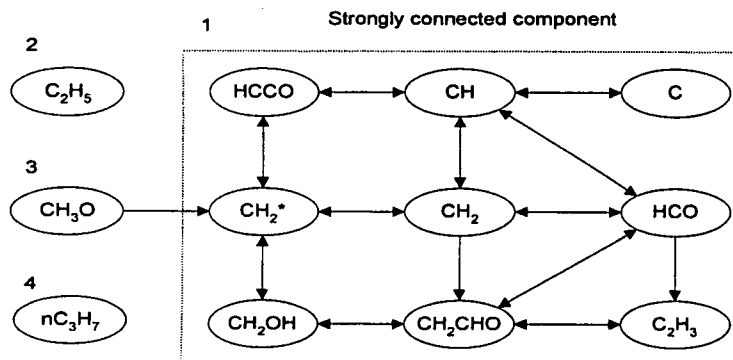

Figure 3

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system was then solved with a directed graph abstracted from the dependence of QSS species. A sample QSSDG for the 16-step ethylene mechanism is shown in Fig. 3.
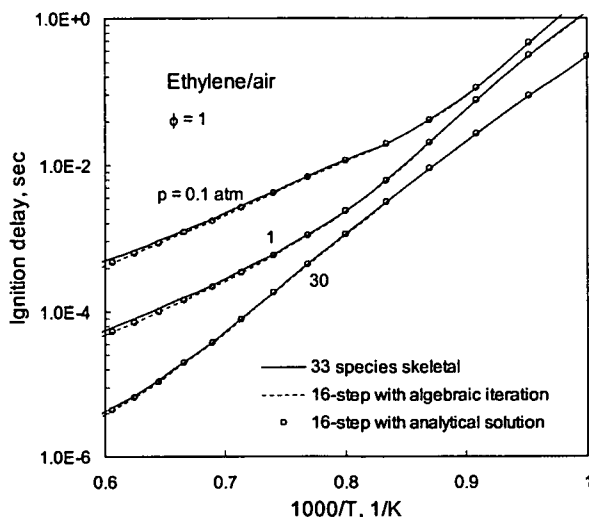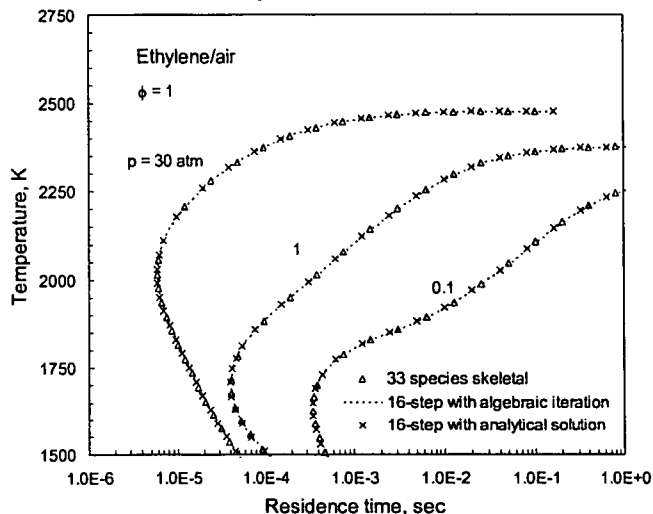

Figure 4

Figure 5

In QSSDG, each node represents a QSS species, and there is a directed edge from QSS species A to B if it directly involves the concentration of B in the QSS expression of A. Strongly connected species form cycles in the graph and result in intrinsically implicit couplings, while couplings between the strongly connected components in QSSDG are acyclic and can be topologically sorted by graph theory. The inner-group couplings were solved efficiently with an algorithm based on eigenvalue analysis of the adjacent matrix of the graph, and the algorithm identifies an optimal or near-optimal sequence for solving the QSS species concentrations within each strongly connected group. The method of QSSDG has been applied to generate a 16-step reduced mechanism for ethylene/air with analytic solution of QSS species concentrations. Good accuracy as well as high efficiency was observed as shown in Figs. 4 and 5 for auto-ignition and PSR respectively, and demonstrates the validity of the linear approximation of the QSS equations.

3 . A reduced mechanism for lean premixed methane/air flames

A reduced mechanism for lean methane/air premixed flames was developed from GRI1.2 using the integrated methods of DRG, DRGASA, and QSSDG. The final reduced mechanism contains 9-steps and 4 QSS species, with the QSS relations solved analytically. The 9-step mechanism was applied to a state-of-the-art 3D DNS simulation for a turbulence Bunsen flame through our collaboration with Sandia National Lab. The solver is explicit 6th-order Rounge-Kutta. Stiffness of the mechanism was removed such that the time step of integration was comparable to that of transport. The simulated flame surface is shown in Figure 6.

Figure 6

4. Extinction and stability analyses of diffusion flames with radiation heat loss

We performed rigorous theoretical analyses on the dual extinction limit behavior and intrinsic flame oscillations near both the extinction states for diffusion flames with radiation heat loss. We first developed a model with the effects of radiation loss in the form of jump relations and reactant leakages across the reaction zone using multi-scale activation energy asymptotics. This model considers properly the excess enthalpy overlooked by previous analyses and incorporates non-unity Lewis numbers of the reactants. It is then applied to study the extinction and stability characteristics of diffusion flames. Figure 7 shows the dependence of fuel leakage, S_F, on the Damköhler number Da^* for different Γ, where Γ is the relative strength of radiation loss to the chemical heat release and the superscript "*" denotes the Damköhler number evaluated at a reference state. It is seen that the solutions are bounded by the two turning points, with the one at smaller Da^*, marked by "■", represents the kinetic extinction limit, $Da^*_{E,K}$, and the one at larger Da^*, marked by "▲", represents the radiative limit, $Da^*_{E,R}$. The neutral stability points, $Da^*_{C,K}$ and $Da^*_{C,R}$, marked by "□" and "△", respectively, indicate that intrinsic flame oscillations develop near both the extinction limits. The flame is unstable within the Damköhler number ranges $Da^*_{E,K} < Da^* < Da^*_{C,K}$ and $Da^*_{C,R} < Da^* < Da^*_{E,R}$. Consequently, steady burning is only possible for $Da^*_{C,K} < Da^* < Da^*_{C,R}$ when flame instability is considered.
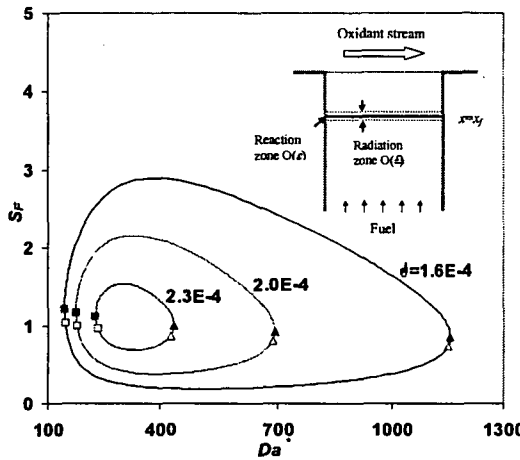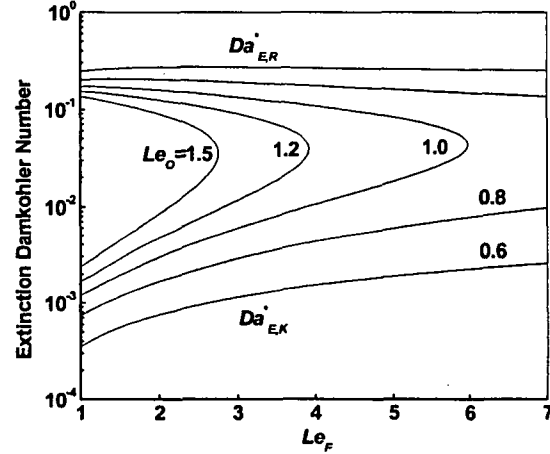

Figure 7

Figure 8

Figure 8 shows variations of the dual extinction Damköhler numbers with the fuel Lewis number, Le_F, for different oxidant Lewis number, Le_O, with fixed Γ. It is seen that the flammable range of Da^* decreases with Le_O and Le_F as it is sufficiently large. When they are both large enough, steady burning is impossible for this value of Γ. With the increase of Le_O, the maximum value of Le_F the system can sustain decreases. Figure 8 indicates that steady burning for the radiative diffusion flames is only possible within a limited range of Lewis numbers. This is because thermal diffusion and radiation are both heat loss mechanisms for flame extinction. Either of them that is large enough is able to induce flame extinction.

Figure 9 shows the normalized unstable range of Damköhler number, $\Omega_R = \left(Da^*_{E,R} - Da^*_{C,R}\right)/Da^*_{E,R}$, versus Le_F for different Γ with $Le_O=1$. It is seen that flame oscillation can indeed develop for Le_F equal to and even well below unity and the unstable range of Le_F is extended with increasing radiation loss. An important result from this analysis is that radiation

loss enhances the heat conduction away from the reaction zone to the fuel and oxidant sides to compensate for the loss, and at the same time suppresses the conduction through the reduction of flame temperature. In general these two opposite effects are not equal and as such radiation loss plays the similar role as varying the thermal diffusivity of the reactants. Therefore, it can be concluded that the imbalance between the thermal and mass diffusions induced by radiation loss under unity Lewis numbers triggers the flame oscillations near the radiation extinction limit. Consequently, the oscillatory instability near the radiation limit can still be considered to be thermal-diffusive in nature.
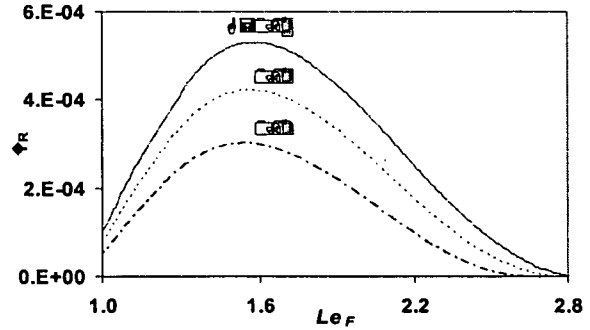
Figure 9

5. Response of premixed flames to velocity fluctuations with flame stretch effects.

To better understand the resonant coupling between unsteady heat release and the natural acoustic modes of combustors, the response of the rate of heat release to velocity fluctuations is studied. Different from previous investigations, the present study considers the variable flame speed due to flame stretch because such a variation is believed to be responsible for some of experimentally observed phenomena. For example, when the Bunsen flame was forced with high frequency, low amplitude disturbances, flame wrinkling was evident only at the flame base and quickly decayed with the axial location farther downstream. Here we only show the results for the effects of flame stretch on the transfer function, defined as the ratio of the normalized fluctuation of heat release rate to that of velocity. In a premixed combustion system, heat release responds to the acoustic modulation through its perturbation to the flame surface area and local burning rate, i.e. flame speed.

Figure 10 shows the spatial distributions of the oscillation amplitude of the flame surface area for different nondimensional flame thickness δ quantifying the extent of flame stretch. The real line corresponds to the case of constant flame speed ($\delta=0$). It is seen that with increasing δ, the flame near the tip region oscillates with increasingly uniform amplitude, implying that flame wrinkling near this region is significantly moderated and the wrinkling is more evident for the flame surface near the burner rim. This explains the experimental observa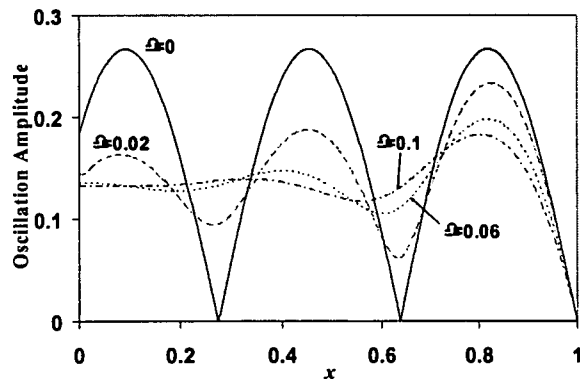tion of more evident flame wrinkling near the flame base. Furthermore, it is seen from the $\delta=0$ case that fluctuation of the flame surface area is a consequence of the superimposition of its string-like oscillation (between nodal points) and bulk oscillatory movement (between the center line and its nearest nodal point). When flame stretch is considered, the nodal points are eliminated so that the flame surface exhibits more bulk movement and less string-like oscillation, which contribute positively and negatively to the fluctuation of flame surface area, respectively.

Figure 10

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